

The phase-in and phase-out of European emissions of HCFC-141b and HCFC-142b under the Montreal Protocol: Evidence from observations at Mace Head, Ireland and Jungfrauoch, Switzerland from 1994 to 2004

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Abstract

The mixing ratios of HCFC-141b (1,1-dichlorofluoroethane) and HCFC-142b (1-chloro-1,1-difluoroethane) have been rising steadily in baseline air at Mace Head, Ireland over the 10-year period from 1994 to 2004. These HCFCs are widely used replacements for the chlorofluorocarbons phased out under the Montreal Protocol and its subsequent amendments. Analysis of the HCFC content of regionally-polluted air arriving at Mace Head from the European continent shows that European emissions reached a peak during 2000–2001 and have declined subsequently, following the phase-out in their usage. European emissions of HCFC-141b have been further constrained by observations at the High-Alpine Jungfrauoch site. The reductions are consistent with the phase-out of HCFC production and use from the year 2001 onwards mandated by European regulations designed to exceed the requirements of the Montreal Protocol.

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1. Introduction

The Montreal Protocol and its subsequent amendments to the Vienna Convention for the Protection of the Ozone Layer has led to the stabilization and slow decay of the global burdens of the chlorofluorocarbons, CFC-11, -12 and -113 (WMO, 2003). The

efficient phase-out of these CFCs has been possible because of the ready availability of replacement substances that can operate within existing equipment and applications. HCFC-141b and -142b have acted as replacement substances for CFC-11, -12 and -113, but because they themselves contain chlorine atoms, they also act as ozone-depleting substances. The Montreal Protocol and its subsequent amendments therefore envisaged that the use of HCFC-141b and -142b would be temporary and that eventually their uses would be phased out (UNEP, 2001).

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HCFC-141b, ($\text{CH}_3\text{CCl}_2\text{F}$), has an atmospheric lifetime of 9.3 years (WMO, 2003) and has been widely employed as a replacement for CFC-11 as a foam blowing agent in the manufacture of rigid polyurethane foams for insulation purposes and of integral skin foams for soles of shoes and motor vehicle dashboards and steering wheels. It has also a use as a replacement for CFC-113 as a solvent for lubricants, coatings and cleaning fluids for aircraft maintenance and electrical equipment. HCFC-142b, (CH_3CClF_2), has an atmospheric lifetime of 17.9 years (WMO, 2003) and has been widely employed as a replacement for CFC-12 as a foam blowing agent for extruded polystyrene board. It has also a small use as a straightforward replacement for CFC-12 in some refrigeration applications (TEAP, 2003). In response to environmental concerns, many countries have begun to reduce consumption of HCFC-141b and -142b to comply with the Montreal Protocol schedule and, in several cases, to make reductions that go beyond compliance with the Protocol. The European Union has regulated for the accelerated phase-out of HCFC consumption, leading to a 75% reduction in HCFC consumption by 1 January 2004 and a complete phase-out by 2010 (European Communities, 2000).

Here we report the continuous, automated, in situ observations of HCFC-141b and of -142b by gas chromatography-mass spectrometry at the Mace Head atmospheric baseline station on the Atlantic Ocean coastline of Ireland. In a previous study, O'Doherty et al. (2004) reported on observations through to June 2002. Here the observations are reported through to December 2004, thereby allowing a complete year to follow the phase-out of HCFC-141b and -142b. By selecting air masses arriving from Europe, we have been able to monitor the levels of HCFC-141b and -142b in pollution events over the 1994–2004 period. This study has been supported by continuous observations of HCFC-141b at Jungfraujoch, Switzerland. Evidence has been found for the phase-in and phase-out of HCFC-141b and -142b usage and emissions, in line with the requirements of the Montreal Protocol and its subsequent amendments.

2. Methodology

2.1. Instrumentation

An automated system for the analysis of atmospheric halocarbons was installed at Mace Head,

Ireland, in October 1994, using an adsorption-desorption unit (ADS) coupled to a Magnum ion trap GC-MS (Finnigan Instruments, Hemel Hempstead, UK) (Simmonds et al., 1995, 1998). The Magnum instrument was replaced in October 1997 with a HP6890 GC coupled to a HP5973 MS (Agilent Ltd., UK). An identical instrument has been operated at Jungfraujoch, Switzerland since 2000. Calibrated measurements of HFC-141b and HCFC-142b have been recorded at Mace Head from 1995 to 2004 and of HCFC-141b at Jungfraujoch from 2000 to 2004.

A full description of the instrument can be found in Prinn et al. (2000) and Sturrock et al. (2001) and a brief summary of the automated GC-MS system is provided here. A nominal, precisely repeatable two litres of ambient air or calibration standard are trapped onto a three-stage mixed composition carbon-based adsorbent contained in a microtrap. The microtrap is located between two thermoelectric coolers maintained at -50°C during sampling. This low temperature enables the small mass of adsorbent material to have the capacity to quantitatively trap the compounds of interest. The trapped sample is purged briefly with clean helium then thermally desorbed to an ultimate temperature of 245°C directly onto a chromatographic capillary column (CPSil-5, 100 m, $5\ \mu\text{m}$ df, 0.32 mm i.d.), interfaced to the Agilent benchtop MS. The individual trace components are separated and identified using both their chromatographic retention times, and their characteristic ion masses. Each component is quantified by integrating the response of a selected target ion and where possible, two qualifier ions are used to ensure compound specific ion ratios are maintained. Custom software developed at Scripps Institution of Oceanography (SIO), enables constant monitoring of ion ratios, peak width ratios and many other diagnostic parameters. This enables rapid identification and determination of instrumental problems. Standard and air analysis are alternated every two hours to provide six calibrated measurements per day.

2.2. Standards and calibration

The method of standard preparation and propagation is described in detail in O'Doherty et al. (2004). Briefly, all working standards are 351 stainless steel electropolished tanks (Essex Cryogenics, Missouri, USA) pressurized with either Mace Head or Cape Grim “real-air”, filled under

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